

Revised Specification

cellulose solution containing cellulose having a degree of polymerization between 200 and 1200 is crosslinked with a saturated hydrocarbon group containing 4 and 16 carbon atoms and said cellulose solution is coagulated to produce a gel that upon dehydration yields a film operable as a battery separator. This separator has higher mechanical strength than uncrosslinked separators.

Detailed Description of the Invention

B1 The present invention discloses a battery separator that improves on the native strength of regenerated cellulose and is more resistant to oxidation than regenerated cellulose. The invention proceeds by crosslinking dissolved cellulose and forming a film from this crosslinked cellulose.

Cellulose, with a degree of polymerization from 200 to 1200, in the form of, but not limited to, microcrystalline cellulose, cotton fiber, paper and microgranular cellulose, is dissolved using a variety of different solvents, including, but not limited to, LiCl/DMAC, trifluoroacetic acid and N-morpholine N-oxide. With LiCl/DMAC, the preferred range is 3 to 8% wt LiCl to DMAC and the applicable range for the percent weight solution of cellulose to solvent is 1 to 11%.

After dissolving the cellulose, the hydroxyl groups on the cellulose are deprotonated by adding an amount of a base, in particular an inorganic hydroxide such as NaOH. The base is added in sufficient quantity to deprotonate just a small fraction of available hydroxyls, usually 1 to 10%, for an excess would result in side reactions causing chain cleavage with consequent depolymerization and degradation in film strength.

After the deprotonation is complete, a dihalide containing 4 to 16 carbon atoms is introduced into the reaction vessel, preferably an alkylene dehalide containing 4-12 carbon atoms. The dihalide reacts with deprotonated sites on adjacent cellulose chains to form saturated hydrocarbon cross-link groups. The cross-link reaction preferably conducted at temperatures from 55 to 90 degrees Celsius for periods ranging from 8 to 24 hours. The amount of dihalide added is the gram-equivalent to all of the hydroxyls available. The NaOH is allowed to settle and the solution is then cast via conventional methods. These methods are known to those skilled in the art of membrane fabrication. They include extrusion of the solution onto a conveyor belt, casting onto a glass plate with a casting knife or casting onto a well-leveled glass plate to form a separator having a thickness from 10 microns to 250 microns.

B2
After casting, the resulting solution is coagulated with conventional techniques, preferably using water as the coagulating agent. Coagulation may be attained either by exposure to ambient moisture or by direct application of a water stream to the resulting solution. The coagulated cellulose material is washed to remove the solvent and the salt. It is possible to employ alcohols mixed with water, but it is preferable that they be kept below 50% by volume.

After thorough washing of the resulting gel, the gel may be dried with any conventional technique such as air drying, vacuum drying or press drying.

Example 1

100 g of LiCl is dissolved in 2 kg of dimethylacetamide (DMAC) at room temperature. 40 g of microcrystalline cellulose (MCC, Aldrich 31,069-7) is placed in a solution containing 2.1 kg of LiCl/DMAC solvent and heated to 120 degrees Celsius for 15 minutes. The

cellulose solution containing cellulose having a degree of polymerization between 200 and 1200 is crosslinked with a hydrocarbon^{Saturated} group containing 4 and 16 carbon atoms and said cellulose solution is coagulated to produce a gel that upon
5 dehydration yields a film operable as a battery separator. This separator has higher mechanical strength than uncrosslinked separators.

Detailed Description of the Invention

10 The present invention discloses a battery separator that improves on the native strength of regenerated cellulose and is more resistant to oxidation than regenerated cellulose. The invention proceeds by crosslinking dissolved cellulose and forming a film from
15 this crosslinked cellulose.

Cellulose, with a degree of polymerization from 200 to 1200, in the form of, but not limited to, microcrystalline cellulose, cotton fiber, paper and microgranular cellulose, is dissolved using a variety of different solvents,
20 including, but not limited to, LiCl/DMAC, trifluoroacetic acid and N-morpholine N-oxide. With LiCl/DMAC, the preferred range is 3 to 8% wt LiCl to DMAC and the applicable range for the percent weight solution of cellulose to solvent is 1 to 11%.

25 After dissolving the cellulose, the hydroxyl groups on the cellulose are deprotonated by adding an amount of a base, in particular an inorganic hydroxide such as NaOH. The base is added in sufficient quantity to deprotonate just a small fraction of available hydroxyls, usually 1 to
30 10%, for an excess would result in side reactions causing chain cleavage with consequent depolymerization and degradation in film strength.

After the deprotonation is complete, a dihalide containing 4 to 16 carbon atoms is introduced into the reaction vessel, ¹ *Preferably an alkylene dihalide containing 4-12 carbon atoms.* The dihalide reacts with deprotonated sites on adjacent cellulose chains to form ^{Saturated} hydrocarbon

5 cross-link groups. The crosslink reaction preferably conducted at temperatures from 55 to 90 degrees Celsius for periods ranging from 8 to 24 hours. The amount of dihalide added is the gram-equivalent to all of the hydroxyls available. The NaOH is allowed to settle and the solution
10 is then cast via conventional methods. These methods are known to those skilled in the art of membrane fabrication. They include extrusion of the solution onto a conveyor belt, casting onto a glass plate with a casting knife or casting onto a well-leveled glass plate to form a separator
15 having a thickness from 10 microns to 250 microns.

After casting, the resulting solution is coagulated with conventional techniques, preferably using water as the coagulating agent. Coagulation may be attained either by exposure to ambient moisture or by direct application of a
20 water stream to the resulting solution. The coagulated cellulose material is washed to remove the solvent and the salt. It is possible to employ alcohols mixed with water, but it is preferable that they be kept below 50% by volume.

After thorough washing of the resulting gel, the gel
25 may be dried with any conventional technique such as air drying, vacuum drying or press drying.

Example 1

100 g of LiCl is dissolved in 2 kg of dimethylacetamide (DMAC) at room temperature. 40 g of
30 microcrystalline cellulose (MCC, Aldrich 31,069-7) is placed in a solution containing 2.1 kg of LiCl/DMAC solvent and heated to 120 degrees Celsius for 15 minutes. The